Authors' response to the review comments

"Synoptic perspectives on pollutant transport patterns observed by satellites over East Asia: Case studies with a conceptual model" by Kim et al.

General responses

The authors express their appreciation to the two reviewers and the editor. We believe that their comments are very productive and substantially contributed to improving the manuscript. We offer general responses and point-by-point responses to the issues and comments addressed by reviewers. Reviewers' comments are shown in italics.

Here are responses for main comments.

(1) Lack of quantitative analysis (Reviewer 1 & 2)

We do understand the reviewers' concern that current manuscript is lack of numerical quantification. It may be our fault because we tried to separate the conceptual approach and numerical approach in current manuscript and concurrent/following studies. In strong agreement with reviewer's comments, we have included more complete model performance evaluations to make this manuscript a standalone research, but also tried to stay in the original scope of the study.

Here are the list of major changes in the manuscript

1. Main model simulations are replaced to the NOAA FNL meteorology-initiated simulations. Both GFS-initiated and FNL-initiated simulations are analyzed.
2. Multiple emission inventories are tested in response to the reviewer's comment on the feasibility of old emission inventories. Two sets of combinations for international and South Korean emission inventories are utilized.
3. We expanded model performance evaluation with further detailed analyses. The additional analyses confirm that model simulations showed very good performance compared to surface observations in their spatial and temporal variation.

In addition, we like to clarify the scope of this study. This study hypothesizes a conceptual model to understand the correlation between regional pollution and synoptic meteorology. We designed this study to suggest control mechanisms of pollution's development and dissipation associated with synoptic systems (and visible evidences from satellites), so it can be further used to design climatological classification over longer term. However, we did not include numerical classification method itself in the current version of the study. Numerical classification, using the conceptual model suggested in this study, is an ongoing next step. As many researchers may agree, the issue of regional air quality in East Asia is very complicated, and we believe that proper classification should be based on conceptual understanding of underlying physics and chemistry.

We do not think that one study can solve all the questions of recent air quality issues in East Asia, especially on the puzzle why East Asia has experienced severe haze events during cold seasons in recent years even though there has been considerable reduction of anthropogenic emissions released from China. This study focuses on the role of synoptic weather among many necessary studies. Hopefully, this study provides a few missing links towards understanding regional air quality and meteorology.

(2) Use of satellite observation (Reviewer 2)

We do not agree that satellites are useless in monitoring regional air quality in East Asia. We understand and agree with the reviewer's concern on the limitation of satellite monitoring, but we also believe in the strong advantages of using satellite data for regional air quality monitoring. Over decades, numerous satellite products have helped monitoring regional pollutants and their precursors.

Satellite monitoring can be very useful because *it can evaluate model outputs and can constrain model inputs*. Satellite product is limited when it was used alone. However, satellite products can provide a good synergy when combined and interpreted with additional information. This study demonstrates the capability of an integrated system of satellite, model and weather analysis to advance understanding in regional air quality. We would not reject the use of satellite data due to its uncertainty.

(3) Scientific importance (Reviewer 2)

In recent years, frequent occurrence of severe haze events in East Asia is one of the most serious public concerns in this region. The reason for the increased haze events is still unknown and very puzzling. Since recent space-borne monitoring of Chinese anthropogenic emissions indicated a decreasing trends in NO_x and SO_2 emissions (Duncan et al., 2016), the role of meteorology has getting more attention, as addressed in the manuscript and the studies mentioned by the reviewer #1. Our manuscript focuses on the possible role of meteorology, especially by the routine passages of synoptic systems, on the formation and removal of regional pollutions in East Asia. This manuscript addresses the importance and governing characteristics of meteorology in the air quality of East Asia.

Duncan et al., 2016: A space-based, high-resolution view of notable changes in urban NOx pollution around the world, doi:10.1002/2015JD024121

We also provide specific responses below.

Anonymous Referee #1

This paper presents an interesting analysis of the relationship between air pollution transport and synoptic weather in East Asia based on weather maps, PM measurements and regional chemical transport modelling. The overall topic of this paper fits to the scope of this journal. However, I think that in the current version the weather typing method was not so objective, and the numerical simulation needs to be improved and validated. Moreover, the discussion part was mainly descriptions on pollution episodes and its associated synoptic weather condition based on horizontal patterns. More quantitative results and in-depth analysis are needed to improve the scientific significance of this work. The paper would benefit from improvements along several main lines, which should amount to major revision.

Thanks for the comment. We apologize that we did not provide more specific the model evaluation previously. We provide more detailed model evaluation in the new manuscript. For example, Figure R1 &R2 show comparison of surface PM_{10}, ozone and NO_2 concentrations during November 2013. Monthly comparisons during all study period (19 months) are included in the supplementary material.

Specific comments:

Conceptual classification of pollutant transport patterns in the work is too subjective. Numerical classification or other objective method is more applicable, especially in terms of pollution forecast.

Thanks for the comment. As mentioned in the general response, this study is designed to provide an intermediate step to advance towards long-term numerical classification. We believe that numerical classification should be designed on the proper understanding of underlying physics and chemistry. In this study, we suggest to go back to the basic and to establish a conceptual model first between systematic occurrence of synoptic patterns and regional pollutant's accumulation and dissipation. Numerical classification, using the conceptual model suggested in this study, is an ongoing next step.

As presented in Section 2, surface weather maps was utilized to determine regional pollutant transport patterns in the present work. However, long-range transport might be more related to 850-hPa and 700-hPa maps, especially for Asian Dust. Please justify why this work used surface maps.

Thanks for the comment. While we agree that 850-mb weather chart is more suited for the analysis of long-range transport of pollutants, especially for the transport of Asian Dust, we have two reasons to provide surface charts in current study. First, unlike Asian dust cases, the transport pathways of anthropogenic pollutants from Chinese cities and industrial areas are not fully understood. Anthropogenic pollutants, especially secondary aerosols, have smaller size distribution compared to Asian dust, and they have higher chance to transport even in lower altitude. We could not exclude the importance of surface synoptic displacement. Second, more practically, most of 850-mb charts are available only for every 12 hours. They did not have enough temporal resolution to be combined with hourly model simulations to analyze detailed transport pathways. For further information, we have included available 850-hPa weather analysis charts in the supplementary materials.

WRF-CMAQ modelling: As described in the Section 3.3, the model was driven by NCEP GFS forecasts data. Here, reanalysis data like FNL with observation assimilation will improve the model performance in meteorology reproduction, and also transport of air pollutants.

We agree that observation-assimilated FNL might provide better meteorology for pollution transport study. Actually, it was a part of our original plan, and we have completed additional simulations using the FNL meteorology during the review process. Results are provided with model performance evaluations. We confirm, however, there is no significant change in our conclusion by the choice of meteorology initiation.

The authors mentioned that the model performance was presented in Kim et al. (2016). However, the modelling time period, meteorological input data and emission inventories used in the present work were all different from those in Kim et al. (2016). Thus, the model evaluation for this simulation (both meteorological parameters and pollutant concentrations) should be conducted and discussed in the manuscript. Observations at 105 surface monitoring sites (Section 4.4) can be used to validate the modelling results.

Kim et al. (2016) was referenced to show the model's physical configurations and general performance. We have included additional model performance evaluations. We apologize for the confusion.

In Section 4, the authors only gave general pictures of horizontal distributions of surface pressure and satellite retrievals using combined plots. It makes no sense to repeatedly show the combined plots throughout the article (Figure 3-8). For the same reason, it's unnecessary to include too much introduction on the geo-referencing method in the manuscript. This section can be substantially improved by more in-depth analysis based on modelling results. Discussion on vertical structure/atmospheric stratification and its impacts on pollutant transport and dispersion will make more sense.

Plots are presented to provide cases of different synoptic patterns corresponding to the classification. We recommend readers to also check animated plots included in the supplementary material since they provide more information on the pollutants' movement.

Geo-referencing technique was mentioned only two times in the main context. Although the technique itself has no importance in a scientific point of view, it is very practical for readers from operational institute. We have received several requests for technical details, including actual code, so they were included in the Appendix and supplementary material.

Satellite observations are excellent for spatial distribution but is technically limited to the evaluation of vertical structure unless they are designed for limb scanning. Figure R3 demonstrates sequences of modeled PM_{10} concentration vertical profiles during Dec. 5-7, 2013 episode, along a cross section over the Yellow Sea. They illustrate the propagation and the vertical extension of intensive pollutant plumes over the Yellow Sea pretty well. Its evaluation, however, is limited in current study. We believe vertical structures can be better analyzed using *in-situ* measurements.

We are also analyzing related vertical structure using *in-situ* aircraft measurements from the 2016 KORUS-AQ campaign. We cannot include any analysis here because they are out of the study period, and also we are not allowed to release any campaign observations before June 2017.

Warm conveyor belts (WCBs), which are associated with cyclones, are crucial in the long-range transport of air pollutants over East Asia. However, in this work, there is no discussion on WCBs. To clarify the relation between synoptic weather and pollutant transport, more focus should to be paid on WCBs.

Thanks for the comment. We included a discussion of the WCB. WCB describes the enhancement of pollutant concentration inside the warm sector – in front of cold front. Initially, we did not use the concept of WCB to avoid unnecessary confusion. While the WCB usually explains strong transport mechanism (both vertically and horizontally), it is quickly followed by strong removal of pollutants due to frontal activities. We have describes the dual role of low pressure systems – transport by WCB and effective removal of local pollutants.

Most part of Section 4 is descriptive, lacking of quantitative analysis and constructive conclusions. The study period is less than two years, leading to the fact that the discussions are more like descriptions on pollution episodes, which has been addressed in detail by many existing studies.

We agree that there have been previous studies to demonstrate pollution episodes, but they are mostly for a short campaign period (compared to 2 year periods in current study), and the use of satellite data is mostly limited. In this study, we introduce more integrated system to demonstrate surface observations, model and satellite observations combined with synoptic weather analysis altogether. For the use of satellite data, there are few studies to demonstrate NO_2 column densities (e.g. as a proxy of anthropogenic emission plumes) from multiple satellites in daily basis. We believe this study provide a very unique platform for regional air quality analysis.

In addition, many conclusions need more support, for instance, the authors concluded that the PM pollution on December 30–31 was caused by anthropogenic emissions and Asian Dust transport without enough evidences. Checking the dust emission rate in the model and observed PM2.5/PM10 ratio can provide more useful information and supportive evidences.

Thanks for the comment. We concur that the $PM_{2.5}$ to PM_{10} mass ratio is a strong indicator for pollutant components, especially for the Asian dust. We expanded the discussion on Dec. 31, 2013 case and included analysis on the change of $PM_{2.5}/PM_{10}$ ratios during the event. Figure R4 shows changes of $PM_{2.5}$,

PM_{10} and theirs ratios observed at the Bulkwang supersite and several Chinese surface sites during the Dec. 31 episode. The $PM_{2.5}$ to PM_{10} ratios were high in the earlier development of the high PM episode, but became lower in the later stage, implying that this case is likely a mixture of anthropogenic pollution (earlier) and Asian dust (later), as described in the manuscript. Observations from Chinese sites also confirm the possibility of Asian dust case, showing lower $PM_{2.5}$ to PM_{10} ratios in northern China. On the other hand, Figure R5 demonstrates a case of strong intrusion of Asian dust (Mar. 18, 2014), showing a high PM_{10} concentration with lower $PM_{2.5}$ to PM_{10} ratios in Bulkwang supersite at Seoul, Korea. Chinese sites also indicate low $PM_{2.5}$ to PM_{10} ratios showing likely Asian dust signal.

Besides, it should be noted that some pollution cases cannot be totally attributed to the transport. Some kinds of synoptic weather pattern might give rise to specific meteorological conditions (strong radiation, high air temperature, less precipitation and stable boundary layer) that favor the formation of secondary pollutions (NO2 and PM) or the accumulation of locally-emitted pollutants. The aforementioned uncertainties should be discussed and clarified here.

Thanks for the comment. We agree with the reviewer's comment that some kinds of synoptic weather pattern is associated with specific meteorological condition that is favorable to the formation of secondary pollution or the accumulation of local pollutants. It is important especially in the case of stagnant higher pressure system where all East Asian countries are under the similar meteorological condition. We already mentioned this point in the conclusion that it is not easy to separate regional transport from local development without a help of a proper chemistry transport modeling. We further clarified the point.

For this point, we have further quantitative analysis to separate the contributions from local and international emission sources to the surface concentration of particulate matter in South Korea (Kim et al., 2017a). Sensitivity to the choice of meteorological model and emission inventories are also discussed in the same study. Indeed, this is more complicated issue if we consider the formation of secondary aerosol during transport. Kim et al. (2017b) also discusses possibility of secondary aerosol formation by precursors from different regional emission sources. Finally, we like to clarify that this study does not have any conclusion on the attribution or responsibility of regional emissions sources to each Asian countries. We better like to understand the control mechanism.

It is actually insignificant to describe the individual pollution episode and its transport pathway in this work. Long-term dataset and statistical analysis could further strengthen the quantitative conclusions and improve the scientific importance.

While simply describing individual pollution episodes may not be important, we did not describe them without reason. We tried to demonstrate how well pollutants are accumulated under high pressure systems, and how effectively low pressure systems sweep out high concentrations. Actually, this finding is connected to our next study (Kim et al., 2017c) that demonstrates that recent increase of surface particulate matter concentration in South Korea is fully explained by the interannual variation of surface wind speed, so the strength of midlatitude ventilation plays a crucial role in regional air quality in East Asia (See Figure R6). This implies that the role of synoptic weather system in East Asia is as large as the rapid change of anthropogenic emissions. We believe current study provides very important link to connect this region's air quality to changes of meteorology and/or climate.

Technical Corrections: Page 5, Line 23: NIER and KMA need to be specified when they are presented for the first time.

Corrected.

Page 6, Line 4-6: surface PM10 concentrations and KMA weather map are introduced twice in Section 3.1 and 3.2.

Corrected.

Section 3.1 is too short to be a section, and the descriptions on graphical technique are redundant.

Section 3.1 is to separate observational and analytical data from satellite and model data. Description on the geo-referencing technique based included based on several requests of operational forecast agencies. If the reviewer still wants to remove it, we will move it to the supplementary material section.

The quality of figures was too bad. The contour labels and legends in figures are not clear enough.

As described in the Appendix, overlapping of weather chart was done pixel-by-pixel graphical transformation, so, technically, it cannot be enhanced beyond its original pixel resolution. Instead, we have included original weather chart graphics files for readers who like to read actual pressure contour labels.

Conclusive remarks

We again appreciate reviewers' comments. We think that most of reviewers' comments are helpful and relevant. We tried our best to cover most of the points addressed by reviewers, and, at the same time, we tried to keep the scope of this study clear and concise. Additional analysis are or will be available in following studies.

1. Quantitative estimation of contributions from regional pollutants and precursors (Kim et al., 2017a)
2. Formation of secondary aerosol (Kim et al., 2017b)
3. Long-term correlation of regional weather and pollution (Kim et al., 2017c)

Kim, E., C. Bae, **H. C. Kim**, J. H. Cho, B.-U. Kim, and S. Kim, 2017a: Regional Contributions to Particulate Matter Concentration in the Seoul Metropolitan Area, Korea: Seasonal Variation and Sensitivity to Meteorology and Emissions Inventory, *Atmospheric Chemistry and Physics Discussion,* doi:10.5194/acp-2016-1114

Kim, B.-U., C. Bae, **H. C. Kim**, E. Kim, and S. Kim, 2017b: Spatially and Chemically Resolved Source Apportionment Analysis: Case Study of High Particulate Matter Event in the Seoul Metropolitan Area, South Korea, during late February 2014, *under review*

Kim, H. C., S. Kim, B.-U. Kim, C.-S. Jin, R. Park, C. Bae, M. Bae, and A. Stein, 2017c: Recent increase of surface particulate matter concentrations in the Seoul Metropolitan Area, Korea, *under reivew*

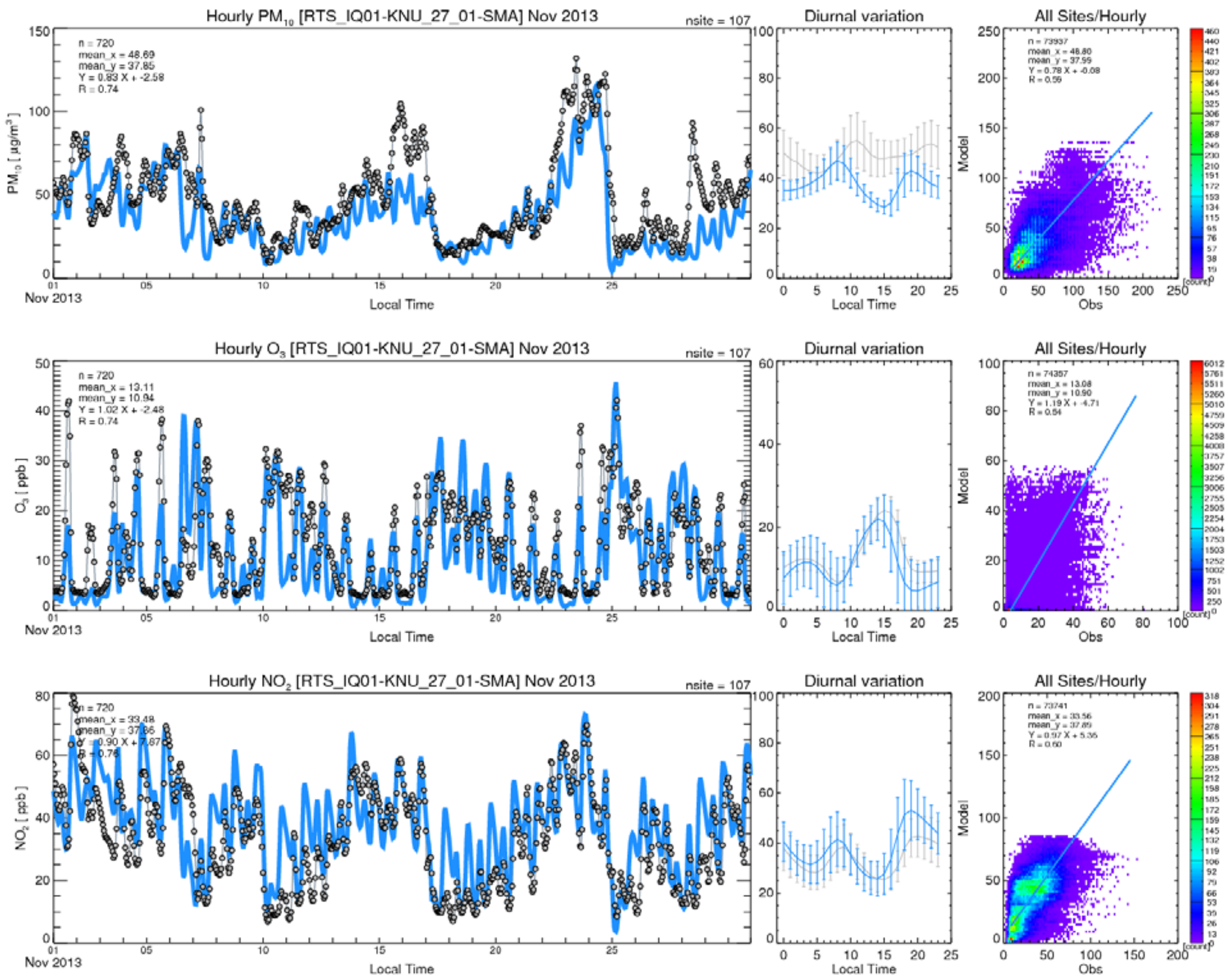

Figure R1 Time series of surface PM10, O3 and NO2 concentrations from model and surface monitoring sites during November 2013 over the SMA, Korea. Observations from 107 sites are compared. Simulation is conducted using the INTEX-B 2006 emission inventory over Asia and CAPSS 2007 over South Korea. Additional comparisons are provided in the supplemental materials.

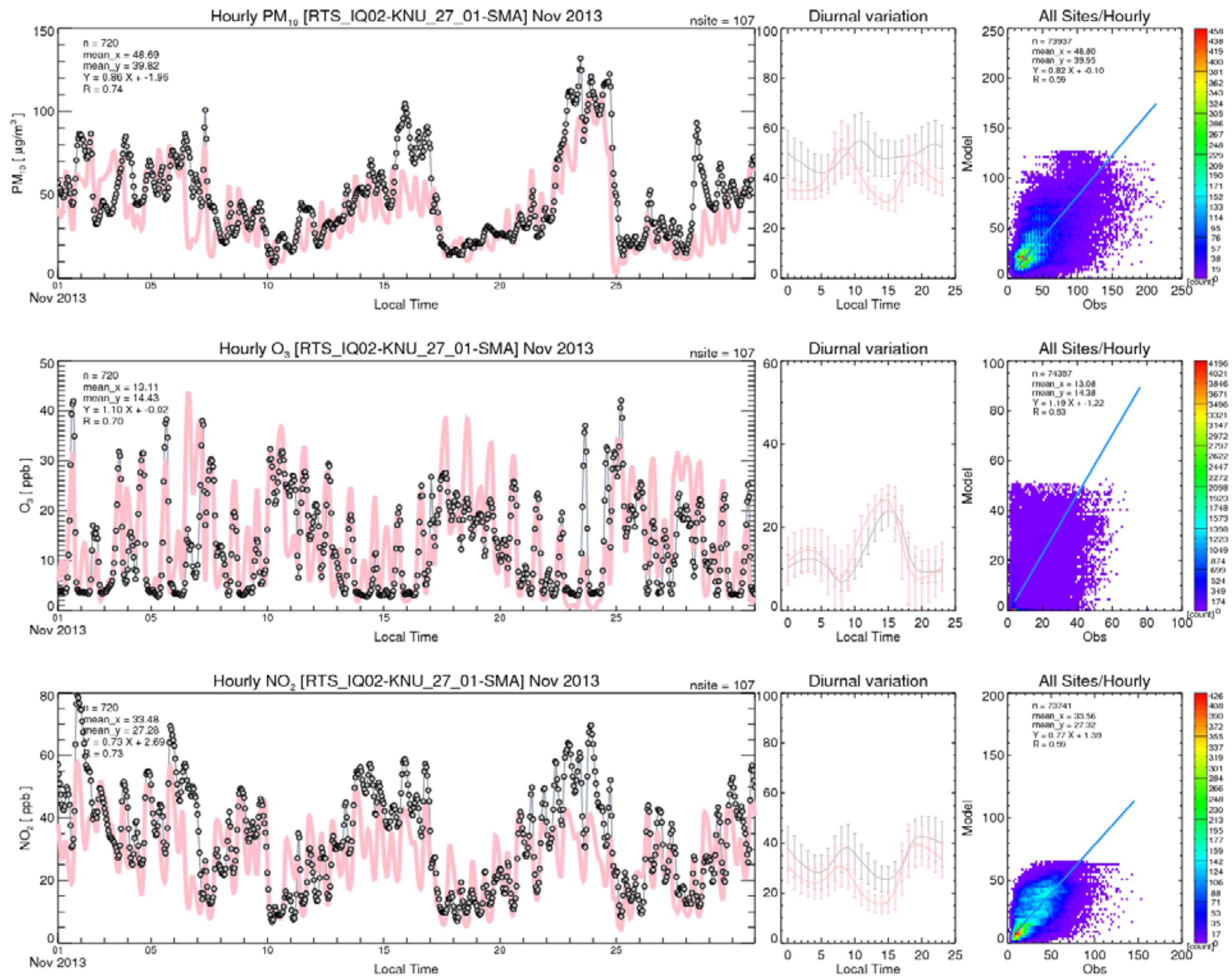

Figure R2 Same with Figure R1 except emission inventories from MICS-Asia 2010 and CAPSS 2010.

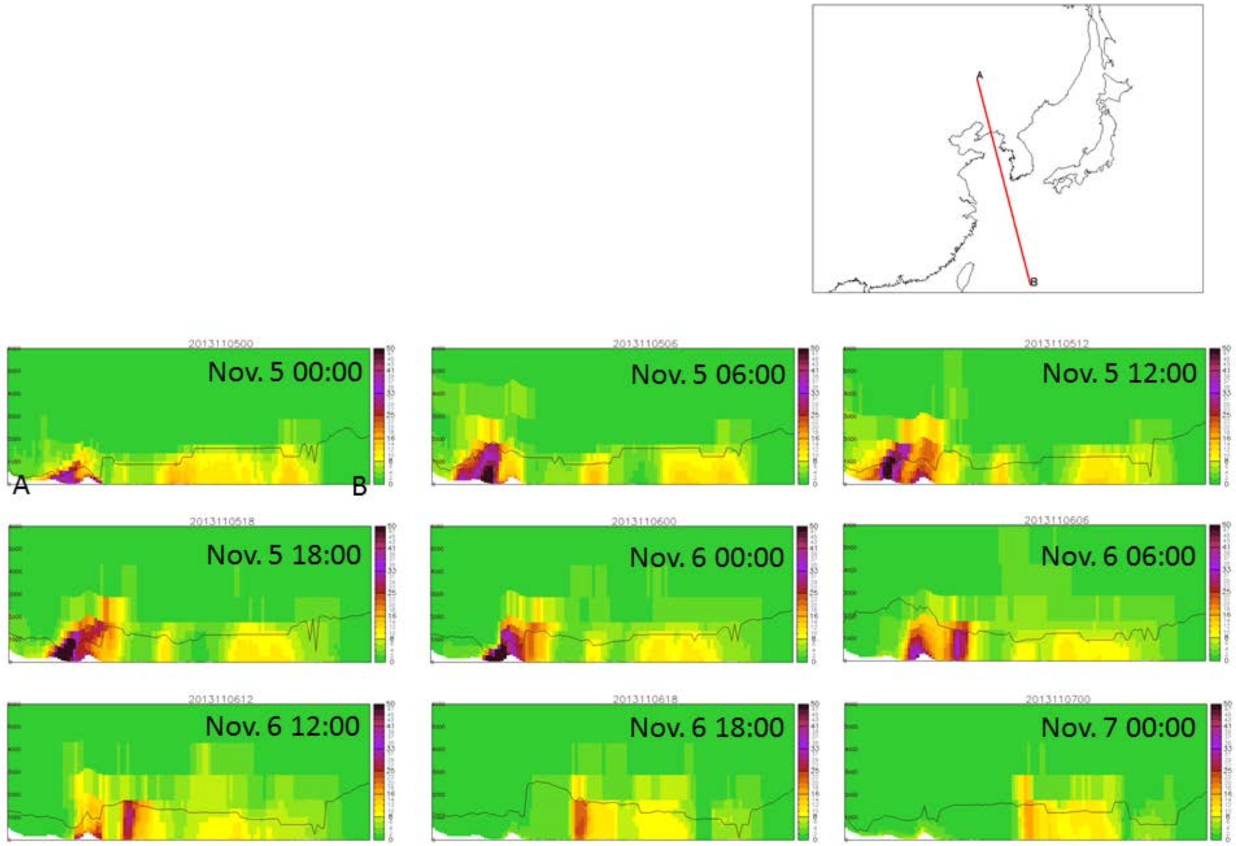

Figure R3 Sequences of PM_{10} concentration vertical profile of cross section over the Yellow Sea (A to B) during Dec. 5-7, 2013. Black lines indicate simulated PBL height.

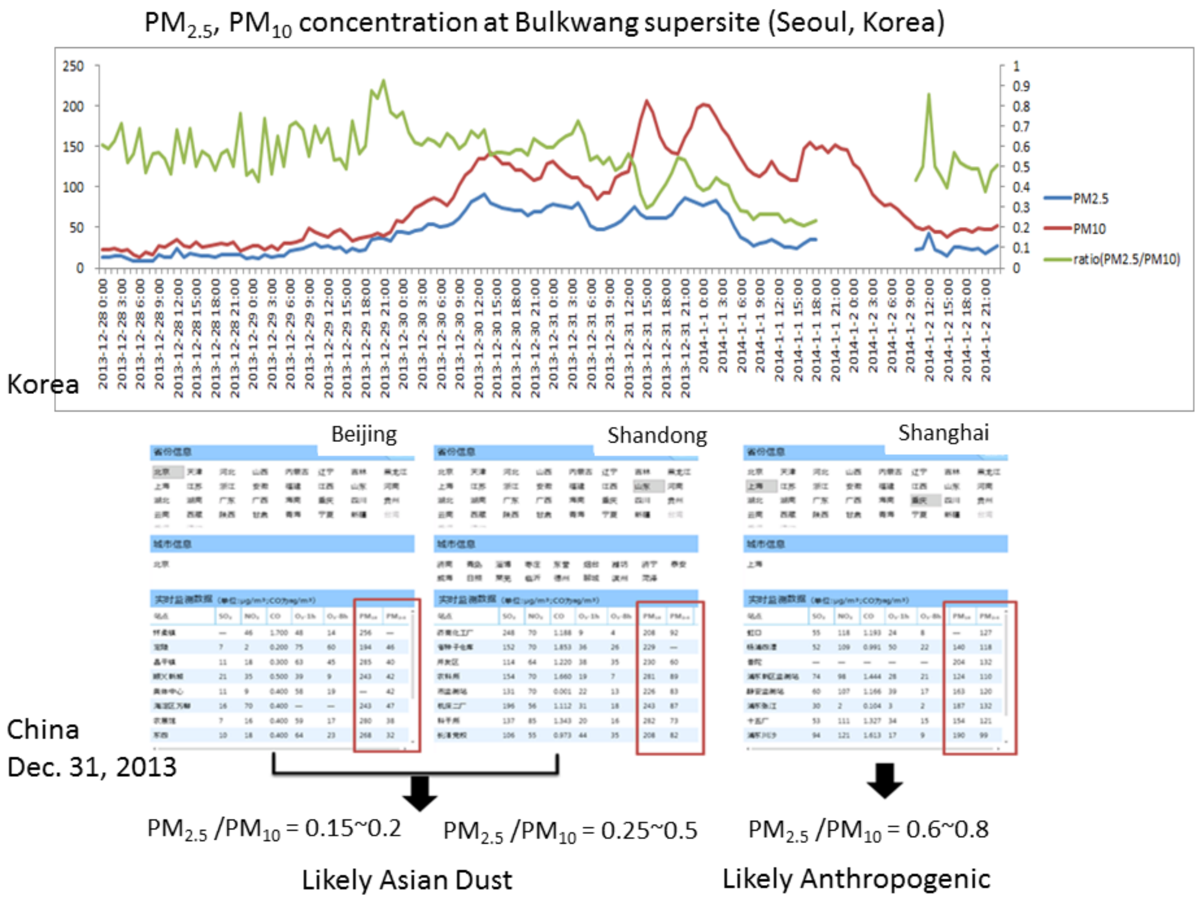

Figure R4 Time series of PM2.5, PM10 and their ratio (PM2.5/PM10) at Bulkwang supersite, Seoul, Korea during Dec. 31, 2013 episode (upper), and monitoring sites at Beijing, Shandong and Shanghai, China (lower).

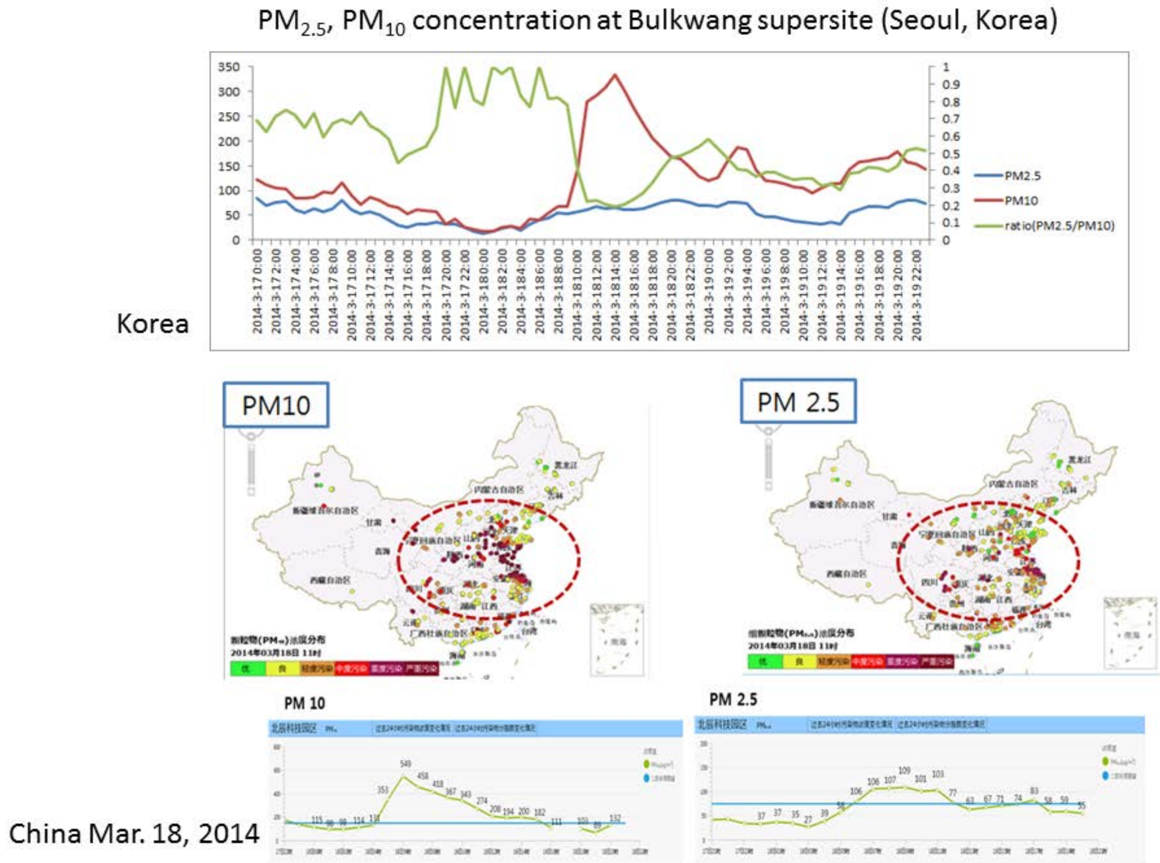

Figure R5 Time series of PM2.5, PM10 and their ratio (PM2.5/PM10) at Bulkwang supersite, Seoul, Korea during Mar. 18, 2014 episode (upper), and monitoring sites at China (lower).

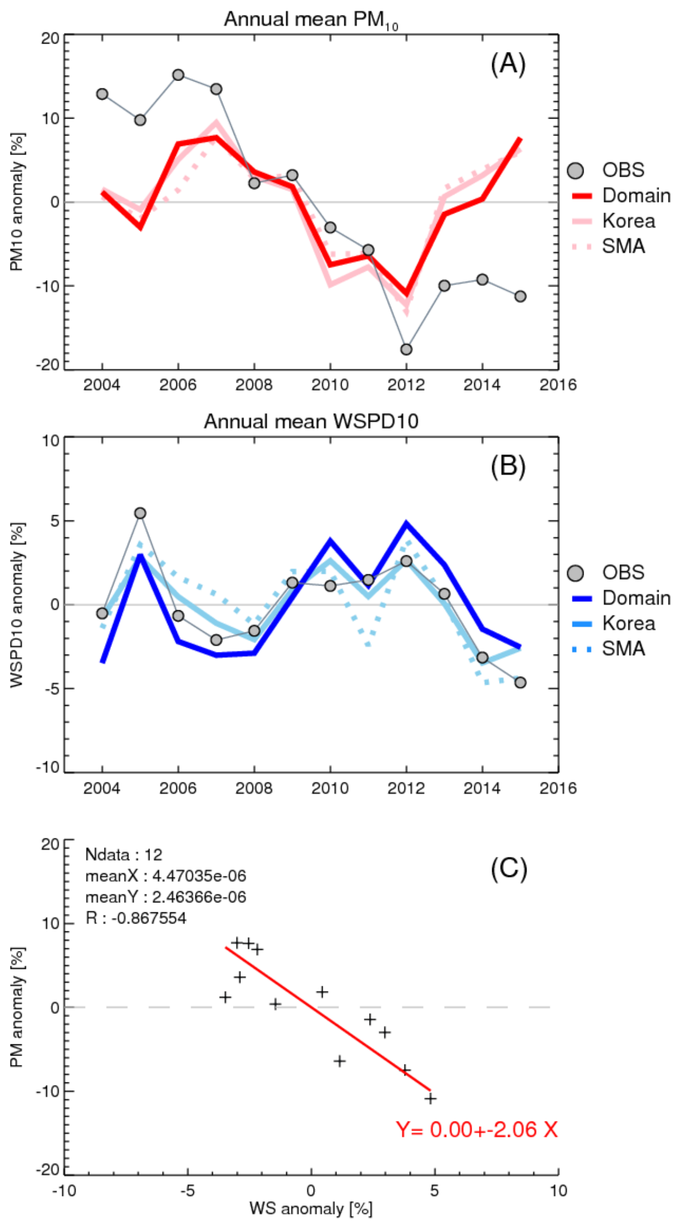

Figure R6. Normalized anomalies of annual mean surface PM concentration (a), and annual mean 10-m wind speed (b). Red (blue), pink (light blue), and dashed pink (light blue) lines indicate anomalies of modeled PM concentrations averaged over the 9-km domain-wide, Korea (land pixels), and the SMA regions for PM concentrations (for wind speed). Circles indicate observations of surface PM concentrations (257 sites over South Korea) and wind speed (79 sites over South Korea). The scatter plot (c) shows a least square regression fit between normalized anomalies of surface PM concentrations and wind speed from the model (9-km domain average). (Kim et al., 2017c)